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Multiple Ionization in Ion-Atom Collisions

Takeshi MUKOYAMA* and Shin ITO†

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The single K- plus multiple L- and M-shell ionization cross sections by light ions have been calculated by the semiclassical approximation. The results are compared with other calculations. The modifications of the straight-line SCA calculations for multiple ionization cross sections are discussed.

KEY WORDS: Multiple ionization/ Inner shell/ Semiclassical approximation/

I. INTRODUCTION

In ion-atom collisions, it is well known that the direct Coulomb interaction between the projectile and the target atom produces the multiple inner-shell vacancies with considerable probabilities. This process is important to obtain information about structures of x-ray and Auger satellites and has been studied extensively.[1] Recently, the multiple ionization process has received a special attention because the intensity of the satellite lines depends on the chemical environments of the target atoms.[2]

The theoretical prediction of the KL^m (single K- and m L-shell) multiple ionization cross sections has been made by Hansteen and Mosebekk[3] in the semiclassical approximation (SCA). Assuming each electron is independent, the cross section is given as the integral over impact parameter of a product of the single ionization probabilities of the atomic shells concerned. McGuire and Richard[4] developed the method to calculate the multiple ionization cross sections in the same manner by the use of the binary-encounter approximation (BEA). McGuire[5] used the BEA to compute KL^m ionization cross sections for various target atoms by alpha particles.

For low projectile energies, both results are in agreement with each other. However, the SCA values become smaller than the BEA ones with increasing the projectile velocity. Hansteen[6] recalculated the KL^1 multiple ionization cross sections in the SCA and his results are larger than the previous SCA values, but still smaller than the BEA values in the high-energy region.

It is th purpose of the present work to calculate the K- plus L- and M-shell multiple ionization cross sections in the SCA and compare with other theoretical predictions.

II. THEORY

If we assume that electrons in the target atom move and interact with the projectile independently with each other, the probability of removing m electrons from

* 向山 毅: Institute for Chemical Research, Kyoto University, Uji, Kyoto, 611 Japan
† 伊藤 眞: Radioisotope Research Center, Kyoto University, Kyoto, 606 Japan

a particular atomic shell containing n electrons by direct Coulomb ionization caused by a heavy charged particle with impact parameter b is given by a binomial distribution[3,4)]

$$\binom{n}{m}\left[\frac{1}{n}I_p(b)\right]^m\left[1-\frac{1}{n}I_p(b)\right]^{n-m} \tag{1}$$

where $I_p(b)$ is the single ionization probability for p shell and $\binom{n}{m}$ is the binomial coefficient.

In the case of K- plus multiple L-shell ionization, the KL^m ionization probability can be written using Eq. (1) as

$$I_{KL^m}(b)=I_K(b)\sum_{rst}\binom{2}{r}\binom{2}{s}\binom{4}{t}\left[\frac{1}{2}I_{L_1}(b)\right]^r\left[1-\frac{1}{2}I_{L_1}(b)\right]^{2-r} \times\left[\frac{1}{2}I_{L_2}(b)\right]^s\left[1-\frac{1}{2}I_{L_2}(b)\right]^{2-s}\left[\frac{1}{4}I_{L_3}(b)\right]^t\left[1-\frac{1}{4}I_{L_3}(b)\right]^{4-t}, \tag{2}$$

where $r+s+t=m$. The total KL^m ionization cross section is calculated by

$$\sigma_{KL^m}=2\pi\int_0^\infty b\,\mathrm{d}b\,I_{KL^m}(b). \tag{3}$$

When the single ionization probability $I_p(b)$ is small, it is the reasonable assumption to set $[1-\frac{1}{n}I_p(b)]\approx 1$. Then Eq. (2) can be simplified as [3,6)]

$$I_{KL^m}(b)=I_K(b)\sum_{rst}\binom{2}{r}\binom{2}{s}\binom{4}{t}\left[\frac{1}{2}I_{L_1}(b)\right]^r\left[\frac{1}{2}I_{L_2}(b)\right]^s\left[\frac{1}{4}I_{L_3}(b)\right]^t, \tag{4}$$

For KL^1, KL^2, and KL^3 ionization, we obtain[7)]

$$I_{KL^1}(b)=I_K(b)I_L(b), \tag{5}$$

$$I_{KL^2}(b)=\frac{1}{2}I_K(b)\left\{I_L^2(b)-\frac{1}{2}\left[I_{L_1}^2(b)+I_{L_2}^2(b)+\frac{1}{2}I_{L_3}^2(b)\right]\right\}, \tag{6}$$

$$I_{KL^3}(b)=\frac{1}{16}I_K(b)\{I_{L_3}^3(b)+4[I_{L_1}(b)I_{L_2}^2(b)+I_{L_1}^2(b)I_{L_2}(b) +I_{L_1}^2(b)I_{L_3}(b)++I_{L_2}^2(b)I_{L_3}(b)]+6[I_{L_1}(b)I_{L_3}^2(b) +I_{L_2}(b)I_{L_3}^2(b)]+16I_{L_1}(b)I_{L_2}(b)I_{L_3}(b)\} \tag{7}$$

where $I_L(b)=I_{L_1}(b)+I_{L_2}(b)+I_{L_3}(b)$.

McGuire and Richard[4)] also derived the similar expressions. However, they neglected the difference in the L subshells and assumed eight equivalent L-shell electrons.

In the case of K- plus multiple M-shell ionization, the ionization probability is given in the manner similar to Eq. (2) as

$$I_{KM^n}(b)=I_K(b)\sum_{rstuv}\binom{2}{r}\binom{2}{s}\binom{4}{t}\binom{4}{u}\binom{6}{v}\left[\frac{1}{2}I_{M_1}(b)\right]^r\left[1-\frac{1}{2}I_{M_1}(b)\right]^{2-r}$$
$$\times\left[\frac{1}{2}I_{M_2}(b)\right]^s\left[1-\frac{1}{2}I_{M_2}(b)\right]^{2-s}\left[\frac{1}{4}I_{M_3}(b)\right]^t\left[1-\frac{1}{4}I_{M_3}(b)\right]^{4-t}$$
$$\times\left[\frac{1}{4}I_{M_4}(b)\right]^u\left[1-\frac{1}{4}I_{M_4}(b)\right]^{4-u}\left[\frac{1}{6}I_{M_5}(b)\right]^v\left[1-\frac{1}{6}I_{M_5}(b)\right]^{6-v}, \tag{8}$$

When the quantity $[1-\frac{1}{n}I_p(b)]$ is nearly equal to unity, the K- plus M-shell multiple ionization probability is written by

$$I_{KM^1}(b)=I_K(b)[I_{M_1}(b)+I_{M_2}(b)+I_{M_3}(b)+I_{M_4}(b)+I_{M_5}(b)] \tag{9}$$

$$I_{KM^2}(b)=\frac{1}{24}I_K(b)\{6[I^2_{M_1}(b)+I^2_{M_2}(b)]+9[I^2_{M_3}(b)+I^2_{M_4}(b)]$$
$$+10I^2_{M_5}(b)+\sum_{\substack{i,j=1\\i<j}}^{5}I_{M_i}(b)I_{M_j}(b)\}\ . \tag{10}$$

III. RESULTS AND DISCUSSION

In the present work, we calculated the single ionization probabilities for K, L and M shells in the straight-line SCA approximation. For this purpose, we used the computer program similar to that developed by Hansteen *et al.*[8] In this case, for the same projectile velocity the single ionization probability is proportional to the square of the projectile charge Z_1. Therefore, we calculated the multiple ionization cross sections only for the case of proton impact on copper.

The ionization probability for the projectile with charge Z_1, mass M_1, and energy E_1 can be obtained from that for proton with energy E_p as

$$I_p(E_1,\ b)=Z_1^2\,I_p^0(E_p,\ b), \tag{11}$$

where $E_p=E_1/M_1$ and $I_p^0(E,\ b)$ is the ionization probability by proton with energy E and impact parameter b.

Table I shows the calculated results of KL^m multiple ionization cross sections for protons on copper. The SCA calculations of Hansteen and Mosebekk[3] and Hansteen[6] are also listed in the table and compared with the present results. Two types of calculations have been made, i.e. with Eq. (2) and with Eq. (4) assuming $[1-\frac{1}{n}I_p(b)]\approx 1$. It can be seen from the table that, in the present case where $I_p(b)$ is small, Eq.(4) is a reasonable approximation to Eq. (2).

The results of Hansteen and Mosebekk[3] and of Hansteen[6] were obtained by the use of Eq. (4). The KL^1 values of Hansteen are in agreement with the present values with Eq. (4). On the other hand, except for the case at 5 MeV, the values of Hansteen

Table I. The K- plus L-shell multiple ionization cross sections for protons on copper (barns).

E (MeV)		σ_{KL^0}	σ_{KL^1}	σ_{KL^2} ($\times 10^2$)	σ_{KL^3} ($\times 10^4$)
0.5	I[a]	3.08	0.104	0.150	0.122
	II[b]	3.18	0.107	0.154	0.124
	HM[c]	2.5	0.060	0.060	
	H[d]	3.4			
1.0	I	24.0	0.930	1.55	1.44
	II	25.0	0.961	1.59	1.44
	HM	23	0.60	0.60	
	H	26	1.1		
2.0	I	121	3.99	5.68	4.58
	II	125	4.10	5.82	4.68
	HM	120	2.40	1.70	
	H	131	4.5		
5.0	I	475	9.21	7.83	3.81
	II	484	9.37	7.95	3.85
	HM	480	12.0	9.30	
	H	481	9.4		
10.0	I	743	7.87	3.66	0.976
	II	752	7.95	3.69	0.983
	HM	740	1.60	0.16	
	H	772	8.1		

[a]Present result with Eq. (2).
[b]Present result with Eq. (4).
[c]Hansteen and Mosebekk, Ref. 3
[d]Hansteen, Ref. 6

and Mosebekk are smaller than the present results.

In Fig. 1, the present results of the KL^m ionization cross sections for protons on copper are plotted against energy and compared with the BEA calculations[4)] and the SCA values of Hansteen and Mosebekk.[3)] The double K-shell ionization cross sections, σ_{K^2}, are also plotted in the figure. As already seen above, the SCA calculations of Hansteen and Mosebekk give smaller cross sections than the present SCA and the BEA.

McGuire and Richard[4)] used the peaked BEA to calculate $I_p(b)$ and obtained the KL^m ionization cross sections by the use of Eq. (2). However, they assumed that eight electrons in the L shell are equivalent. The KL^m ionization probability in this approximation is simplified as

$$I_{KL^m}(b)=I_K(b)\binom{8}{m}\left[\frac{1}{8}I_L(b)\right]^m\left[1-\frac{1}{8}I_L(b)\right]^{8-m}. \tag{12}$$

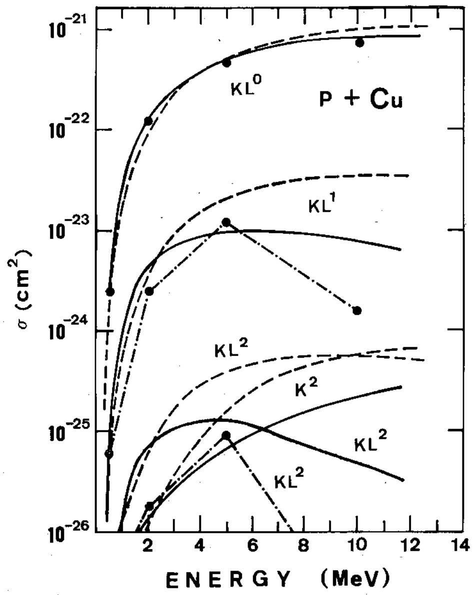

Fig. 1. K- and L-shell multiple ionization cross sections for protons on copper. The solid curves represent the present results, the dashed curves indicate the BEA calculations of McGuire and Richard (Ref. 4), and the solid circles and the dot-dashed curves are the SCA values of Hansteen and Mosebekk (Ref. 3).

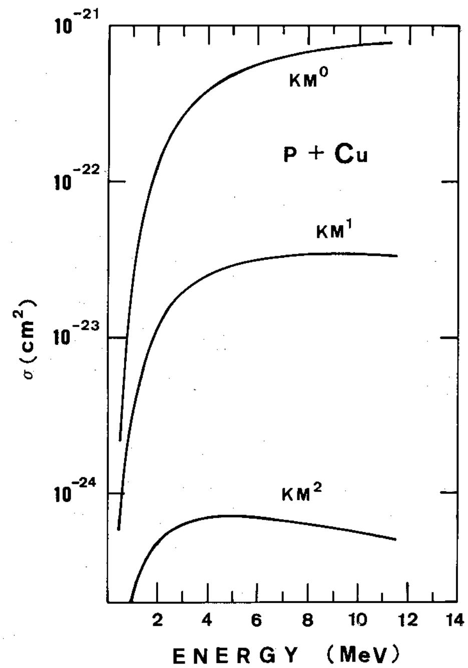

Fig. 2. K- plus multiple M-shell ionization cross sections for protons on cupper.

The present SCA results agree well with the BEA in the low-energy region, but decrease more rapidly with increasing energy. In the straight-line SCA and the BEA, the inner-shell ionization cross section reaches the maximum at $E/\lambda U \approx 1$ and decreases with projectile energy, where E is the projectile energy, λ is the ratio of the projectile mass to that of the electron and U is the binding energy of the target electron. According to this fact, the K-shell ionization cross section for protons on copper increases with energy in the energy region of Fig. 1, but the L-shell ionization cross section has a maximum about at 2 MeV and decreases for higher energies. Considering the energy dependence of the K- and L-shell ionization cross sections, the decrease in the KL^m ionization cross section in the high-energy region is quite reasonable. McGuire[5] pointed out that their KL^m ionization cross sections in the BEA may be overestimated for low Z because $I_p(b)$ is probably too large.

Figure 2 shows the KM^m multiple ionization cross sections for protons on copper in the SCA. In this case, the maximum value for the M-shell ionization cross sections is located at very low energies, less than 0.2 MeV, and the M-shell ionization cross sections decrease gradually in the whole energy range considered here, although the K-shell ionization cross section increases.

When the projectile energy is low, the K-shell ionization takes place mostly near the target nucleus, i.e. at small impact parameters. Since the L- and M-shell ionization probabilities are slowly varying in this region, $I_p(b)$ for L- and M-shell ionization can be approximated by $I_p(0)$ and taken outside the integral in Eq. (3). The integration over impact parameter is carried out only for $I_K(b)$ and the KL^m multiple ionization cross section is approximated by

$$\sigma_{KL^m} = \sigma_K \sum_{rst} \binom{2}{r}\binom{2}{m}\binom{4}{t} \left[\frac{1}{2} I_{L_1}(0)\right]^r \left[1-\frac{1}{2} I_{L_1}(0)\right]^{2-r} \times \left[\frac{1}{2} I_{L_2}(0)\right]^s \left[1-\frac{1}{2} I_{L_2}(0)\right]^{2-s} \left[\frac{1}{4} I_{L_3}(0)\right]^t \left[1-\frac{1}{4} I_{L_3}(0)\right]^{4-t}, \tag{13}$$

where σ_K is the K-shell ionization cross sections.

In order to use Eq. (13), it is important to know that the impact parameter b for K-shell ionization is small. This can be tested by calculating the mean impact parameter $<b>$ for K-shell ionization. Lapicki and Losonsky[9] proposed to use the expression for the mean impact parameter

$$<b> = A_0/q_0, \tag{14}$$

where $q_0 = \triangle E/v_i$ is the minimum momentum transfer for ionization, $\triangle E$ corresponds to the binding energy of the target electron, and A_0 is the constant. For K-shell ionization, A_0 is chosen to be 0.85, $A_0 = 1.5$ for s-state ionization and 2.0 for p-state ionization.

We estimated the mean impact parameter $<b>$ for K-shell ionization of copper atom by proton impact as

Table II. The mean impact parameter for K-shell ionization for protons on copper (a.u.).

E (MeV)	$\langle b \rangle$	$0.85/q_0$
0.5	0.0205	0.0115
1.0	0.0277	0.0163
2.0	0.0352	0.0230
5.0	0.0477	0.0364
10.0	0.0568	0.0515

$$\langle b \rangle = \int_0^\infty I_K(p) b^2 \mathrm{d}b \Big/ \int_0^\infty I_K(b) b \mathrm{d}b \tag{15}$$

The results are listed in Table II and compared with the prediction by Eq. (14). It can be seen that Eq. (14) underpredicts $\langle b \rangle$ values for low-energy projectiles. If we choose $A_0 = 1.5$ for K-shell ionization, the $\langle b \rangle$ values for Eq. (14) are in agreement with those from Eq. (15) in the low-energy region, but become too large for high energies.

For the region of the $\langle b \rangle$ values in Table II, the L-shell ionization probabilities for protons on copper change slowly with b and the difference between $I_p(0)$ and $I_p(\langle b \rangle)$ in Eq. (13) is less than 20%. This fact suggests that we can use $I_p(0)$ or $I_p(\langle b \rangle)$ in Eq. (13) for estimation of the KL^m multiple ionization cross section.

For projectiles with large Z_1, the approximation $[1-\frac{1}{n} I_p(b)] \approx 1$ is not valid, because the $I_p(b)$ values are large. In this case, we must use Eq. (2) to calculate the multiple ionization cross sections. In addition, for highly-charged projectiles, the ionization probabilities in the SCA and the BEA sometimes exceed unity due to the Z_1^2 scaling law and violates the unitarity condition. This fact means that the simple SCA or BEA theory cannot be used to estimate $I_p(b)$. In such a case, the multiple ionization probabilities should be obtained by the coupled-channel calculations[10] or the geometrical model.[11-13]

When the degree of multiple ionization is large, the binding energies of the target electrons change during the ionization process and a simple binomial expression discussed above gives smaller cross sections for highly-ionized states. If we assume that all L and M electrons are ejected *simultaneously*, we can take into account this effect by modifying the binding energies and using an average binding energy for multiply charged states in Eq. (2). On the other hand, if the multiple ionization process takes plase *sequentially*, the ionization probability is no longer expressed by the binomial distribution. Watson *et al.*[14] derived the expression for the distribution of multiple ionization in such a case.

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